

REMARKS

For the Examiner's convenience, the arguments made in the amendment filed April 28, 2003 are repeated below.

The rejection of Claims 1-20 under 35 U.S.C. §112, first paragraph, is respectfully traversed. Indeed, the rejection is now moot in view of the above-discussed amendment. Accordingly, it is respectfully requested that this rejection be withdrawn.

In the Office Action, the Examiner stated that if the above-discussed amendment were made, the claims would be subject to the prior art rejections of record. Since the Examiner has not responded to the arguments on the merits of these rejections in the last response, they will be repeated and expanded below. Even without the term "having no terminal amino group," it is respectfully submitted that the claims are not unpatentable over the applied prior art as now discussed.

The previous rejections of:

Claims 1-2 and 6-5 [sic] under 35 U.S.C. §102(b) as anticipated by or, in the alternative, under 35 U.S.C. §103(a) as obvious over, U.S. 5,349,023 (Ikeda et al); and

Claims 1-10 under 35 U.S.C. §103(a) as unpatentable over Ikeda et al in view of U.S. 5,712,334 (Watanabe et al) taken with U.S. 4,492,783 (Tanaka et al) or the abstract of JP405163-62A (Tsuboi et al),

are respectfully traversed.

The present invention relates to a ceramic-molding binder, and a compression-molding binder for ceramics, and ceramic molding compositions thereof, respectively, each comprising a particular vinyl alcohol polymer. The present invention also relates to a method of producing a ceramic molding.

As described in the specification under "Description of the Related Art," beginning at page 1, second paragraph, there has been a demand in the art for ceramics having better physical, thermal, electrical, mechanical properties and the like, in various shapes and sizes. Smaller moldings with more complex shapes have been accompanied with problems occurring during the handling of green moldings, which are unsintered moldings, including A) greater molding difficulties and defects in portions of green moldings, B) greater difficulty handling green moldings which are far more brittle, and C) cracks in ceramic moldings after the green moldings have been sintered. The amount of organic binder that is added has conventionally been increased to deal with such problems. However, increasing the amount of binder results in other problems, including 1) problems removing the binder, 2) problems with contamination by impurities, etc., and 3) problems with dimensional precision. Various vinyl alcohol polymers have been suggested to address these problems. While the known binders can be successfully used in small amounts, have good miscibility, and are better than conventional binders in terms of performance such as strength, they do not provide satisfactory performance when forming smaller products of more complex shape, particularly by compression-molding.

The presently-claimed invention addresses these problems. Particularly, the ceramic-molding binder of the invention can be added in lower amounts to starting material powder so as to avoid the aforementioned problems 1) through 3), without any of the aforementioned problems A), B), and C).

As recited in the claims, the vinyl alcohol polymer (PVA) of the present invention has an ethylene unit content of 2 to 19 mole %, a polymerization degree of 200 to 2,000, a degree of saponification of 80 to 99.99 mole %, a total content of carboxyl group and lactone rings of

0.02 to 0.4 mole %, wherein the carboxylic acid and lactone ring content in the vinyl alcohol polymer satisfies the following Formula I:

$$-1.94 \times 10^{-5} \times P + 0.044 \leq \text{content} \leq -1.39 \times 10^{-4} \times P + 0.42 \text{ (I)}$$

(where the content (in mole %) represents the content of carboxylic acid and lactone rings, and P represents the viscosity average degree of polymerization of the vinyl alcohol polymer).

Applicants have provided a wealth of comparative data in the specification demonstrating the superior results when operating within the above-recited ethylene unit, polymerization degree, a degree of saponification, and total content of carboxyl group and lactone rings, ranges of the claims.

PVA Polymers A through L are according to the present invention. In view of the above amendment, only PVA Polymers A through K are according to the present invention; PVA Polymers K and L are now for purposes of comparison. Additionally, PVA Polymers M through W, and X through AF are for purposes of comparison. Table 1, at page 23 of the specification, details the characteristics of PVA Polymers A-L. Table 2, at page 24 of the specification, details characteristics of PVA Polymers M-W. Table 3, at page 25 of the specification, details the characteristics of PVA Polymers X-AF. Tables 1, 2 and 3 are reproduced below:

Table 1

Type of PVA polymer	Vinyl alcohol units (mole %)	Vinyl ester units (mole %)	Ethylene units (mole %)	Carboxylic acid and lactone units (mole %)	Degree of Saponification	Polymerization degree
A	94.0	1.4	4.5	0.05	98.5	1550
B	90.0	5.4	4.5	0.05	94.3	1550
C	93.9	1.5	4.5	0.06	98.4	1320
D	88.8	6.6	4.5	0.06	93.1	1320
E	93.1	1.3	5.5	0.10	98.6	540
F	92.0	0.7	7.0	0.30	99.2	510
G	84.0	11.3	4.5	0.15	88.2	1480
H	95.0	1.4	3.5	0.05	98.5	1950
I	91.0	1.7	7.0	0.30	98.2	330
J	85.4	0.7	13.6	0.30	99.2	280
K	85.3	11.3	3.0	0.35	88.3	1320
L	89.8	1.6	8.5	0.03	98.2	280

Table 2

Type of PVA polymer	Vinyl alcohol units (mole %)	Vinyl ester units (mole %)	Other monomer units (mole %)	Degree of Saponification	Polymerization degree
M	88.0	12.0	None	88.0	550
N	98.5	1.5	None	98.5	550
O	75.5	24.5	None	75.5	550
P	88.0	12.0	None	88.0	1750
Q	98.5	1.5	None	98.5	1750
R	92.3	0.7	Veova -10 ^{a)} 4.0 Itaconic acid 3.0	99.2	460
S	60.3	29.5	Allyl acetate 10.2	67.1	256
T	66.7	29.8	Allyl acetate 3.5	66.0	310
U	69.6	13.9	5-hexene-1,2-diol 16.5	83.4	265
V	58.6	25.0	5-hexene-1-ol 15.4 Itaconic acid 1.0	70.1	285
W	67.0	23.5	Ethylene 9.2 AMPS ^{b)} 0.3	74.0	295

(a) Velva-10: total C₁₀ branched fatty acid vinyl ester (*SHELL KAKAGU*)

(b) AMPS: 2-acrylamidopropylsulfonic acid

Table 3

Type of PVA polymer	Vinyl alcohol units (mole %)	Vinyl ester units (mole %)	Ethylene units (mole %)	Carboxylic acid and lactone units (mole %)	Degree of Saponification	Polymerization degree
X	94.7	0.7	2.5	2.05	99.2	2400
Y	94.1	2.8	3.0	0.01	97.0	150
Z	73.2	23.7	1.5	1.54	75.5	500
AA	97.2	1.5	1.2	0.05	98.5	1500
AB	72.3	23.9	3.5	0.3	75.2	500
AC	90.9	1.4	4.5	0.2	98.5	150
AD	95.5	1.4	3.5	0.03	98.5	2400
AE	96.5	0.4	3.5	0.01	99.5	1500
AF	92.9	0.5	4.5	2.05	99.5	1500

In addition, above Table 1 has been modified to include columns for " $-1.94 \times 10^{-5} \times P + 0.044$ " and " $-1.39 \times 10^{-4} \times P + 0.42$ ":

Type of PVA polymer	Carboxylic acid and lactone units (mole %)	Polymerization degree	$-1.94 \times 10^{-5} \times P + 0.044$	$-1.39 \times 10^{-4} \times P + 0.42$
A	0.05	1550	0.014	0.21
B	0.05	1550	0.014	0.21
C	0.06	1320	0.018	0.22
D	0.06	1320	0.018	0.22
E	0.10	540	0.034	0.35
F	0.30	510	0.034	0.35
G	0.15	1480	0.015	0.21
H	0.05	1950	0.006	0.15
I	0.30	330	0.038	0.37
J	0.30	280	0.039	0.38
K	0.35	1320	0.018	0.22
L	0.03	280	0.039	0.38

The above PVA polymers were employed in examples (according to the invention) and comparative examples (for purposes of comparison). Examples 1-10 employ PVA polymers A-J, respectively. (Comparative) Examples 11 and 12 employ PVA polymers K-L, respectively. Comparative Examples 1-20 employ PVA Polymers M-AF, respectively. The examples and comparative examples were prepared as described for Example 1 at page 26, line 10ff. The examples and comparative examples were evaluated for (1) molding strength (toughness); (2) formability; (3) handling properties of moldings; and (4) ceramic moldings, as described in the specification at page 27, line 17, through the end of page 9. The results are shown in Tables 4A and 4B, at pages 30 and 31, respectively, of the specification. A copy of Tables 4A and 4B is **attached herewith**.¹

Applicants describe the data at page 32, line 1, through page 34, line 2, as follows:

The results in Tables 4A and 4B show that the use of the ceramic-molding binder of the present invention resulted in strength equal to or greater than that obtained with the use of conventional binders when forming green moldings having a relatively simple shape. Particularly during the formation of small green moldings of complex shape, not only was the green formability (compressing properties and mold staining) better, but the resulting green moldings also had better handling properties. There were also far fewer cracks in ceramic sinters obtained upon the sintering of the green moldings, resulting in better yields.

Although the use of conventional vinyl alcohol polymers as binders did improve the strength of green moldings of relatively simple shape (Comparative Examples 6 through 11), efforts to obtain small green moldings of more complex shape did not result in altogether favorable granule pressure propagation unless a certain level of molding pressure was applied. The tips of the fine areas of the moldings in particular were found to suffer from poor compressing properties. Although higher molding pressure resulted in some improvement in compressing properties, there was also more obvious staining of the molds. It was thus impossible to simultaneously achieve satisfactory compressing properties while avoiding mold stains. The handling properties of the green moldings were also not altogether satisfactory. More cracks

¹In view of the above-discussed amendment, Examples 11 and 12 in Table 4A, which use PVA Polymers K and L, respectively, are now comparative examples.

were also found in the sintered ceramic moldings, making it impossible to achieve satisfactory effects (Comparative Examples 1 through 11). Even the use of PVA polymers having ethylene units did not afford satisfactory results (Comparative Examples 12 through 20) if the ethylene units, the carboxylic acid and lactone units, the degree of saponification, and the polymerization degree were outside the ranges stipulated in the present invention.

In contrast, the use of the ceramic-molding binder of the present invention simultaneously afforded satisfactory compressing properties while avoiding mold stains, gave green moldings with satisfactory handling properties, and resulted in higher yields of sintered ceramic moldings. These properties were clearly better than those obtained with the use of conventional binders.

Compared to conventional binders, the ceramic-molding binder of the present invention affords better formability and green moldings with better handling properties, particularly during the formation of smaller products of complex shape in various molding methods, especially compression-molding. Ceramic moldings obtained by sintering green moldings using the ceramic-molding binder of the present invention have far fewer cracks and better yields. The present invention thus provides the greater performance demanded of ceramic products, such as better quality.

The above-discussed results could not have been predicted by the applied prior art.

Ikeda et al disclose a vinyl alcohol copolymer having a terminal amino group, comprising 1 to 90 mole % of vinyl alcohol units, 0 to 89 mole % of vinyl esters units and 10 to 90 mole % of units from an ethylenically unsaturated monomer copolymerizable with the vinyl ester (column 2, lines 14-20) wherein the ethylenically unsaturated monomer may be, *inter alia*, ethylene (column 3, line 56). Ikeda et al disclose further that "[w]here units from a monomer having carboxylic group or functional group convertible to carboxylic group or having ester bond such as lactone ring are present in the main chain other than the end of the copolymer having vinyl ester units, their content in the copolymer having vinyl ester units is smaller the better, since they may possibly cause amino groups to be introduced into the side chains of the monovalent copolymer. Thus, the content of units from such a monomer is preferably less than 0.5 mol %, more preferably less than 0.3 mol % and most preferably less than 0.1 mol %" (column 5, lines 49-60).

It is thus clear that Ikeda et al seeks to limit, if not exclude, carboxylic groups and lactone rings because they may cause amino groups to be introduced into side chains; may but do not require ethylene units; disclose nothing regarding polymerization degree; and contains monomer amount ranges amounts and degree of saponification amount ranges significantly greater than the corresponding ranges of the present claims.

In addition, in Ikeda et al, a polymerization level of the vinyl alcohol copolymer is disclosed in term of intrinsic viscosity. Ikeda et al does not disclose a polymerization degree, as recited in the present claims. Formula I of the claims defines the preferred range of the content of carboxylic acid and lactone rings with respect to polymerization degree. Ikeda et al discloses and suggests nothing regarding satisfying said Formula I.

Ikeda et al does not anticipate the presently-claimed invention. As stated in *In re Arkley*, 172 USPQ 524, 526 (CCPA 1972) (copy of record):

[R]ejections under 35 U.S.C. 102 are proper only when the claimed subject matter is identically disclosed or described in "the prior art." Thus, for the instant rejection under 35 U.S.C. [102(b)] to have been proper, the . . . reference must clearly and unequivocally disclose the claimed [subject matter] or direct those skilled in the art to the [subject matter] without any need for picking, choosing, and combining various disclosures not directly related to each other by the teachings of the cited reference. Such picking and choosing may be entirely proper in the making of a 103, obviousness rejection, where the applicant must be afforded an opportunity to rebut with objective evidence any inference of obviousness which may arise from the similarity of the subject matter which he claims to the prior art, but it has no place in the making of a 102, anticipation rejection.

Even picking and choosing in Ikeda et al would not result in the presently-claimed invention. Nor does Ikeda et al render the presently-claimed invention *prima facie* obvious, since without the present disclosure as a guide, one skilled in the art following Ikeda et al would attempt to eliminate carboxyl groups and lactone rings altogether, and would appreciate nothing regarding polymerization degree and the required presence of ethylene units, let alone within the presently-recited ranges, and satisfying Formula I. Moreover, even if a *prima facie* case of obviousness had been made out, the above-discussed comparative data overcomes it.

Specifically, PVA Polymers M through Q contain no carboxylic acid or lactone ring components, and thus represent the preferred goal of Ikeda et al. These PVA Polymers are used in Comparative Examples 1-5, respectively. As Table 4B shows, these comparative examples were substantially inferior to the examples of the invention in every one of the evaluations. PVA Polymer AE meets all the terms of the present claims, except that the content of carboxylic acid and lactone ring is 0.01 mol%. Yet corresponding Comparative Example 19, as shown in Table 4B, had generally inferior compressing and handling properties compared to the examples. PVA Polymer AF meets all the terms of the present claims except that it has a carboxylic acid and lactone ring content of 2.05 mol%. Yet corresponding Comparative Example 20 had not only inferior compressing and handling properties compared to the examples, but was substantially worse in terms of mold staining as well. Similar analyses can be carried out, with regard to the claimed invention, as follows: differences in ethylene unit content (PVA Polymer AA and Comparative Example 15); degree of saponification (PVA Polymer AB and Comparative Example 16); polymerization degree (PVA Polymers AC and AD and Comparative Examples 17 and 18, respectively). Additional

analyses can be made with regard to PVA Polymers R through W, corresponding to Comparative Examples 6-11, wherein other monomer units are present, as shown in Table 2.

Clearly, even with the disclosure of Ikeda et al as a guide, one skilled in the art could not have predicted the above-discussed results, and the influence of the various parameters recited in the claims on these results.

Watanabe et al disclose a lactone-modified polyvinyl alcohol, and a lactone-modified resin having hydroxyl groups which can be derived from a polymer containing 10-5,000 hydroxyl groups, which polymer may be, *inter alia*, an ethylene oxide-modified polyvinyl alcohol (column 18, line 37 through column 19, line 5). In the preparation of the lactone-modified PVA of Watanabe et al, a lactone ring-opening reaction occurs when the polymer having hydroxyl groups reacts with the lactone monomer, whereby the obtained lactone-modified PVA has no lactone ring (see the general formula (1) at column 6).

Watanabe et al taken with Ikeda et al does not render the presently-claimed invention *prima facie* obvious. The lactone-modified polyvinyl alcohol and lactone-modified resin having hydroxyl groups do not appear to contain ethylene units. Nor would the amount of lactone rings appear to be within the presently-recited range. Indeed, the PVA of the present invention has a carboxylic acid and lactone ring content of 0.02 to 0.4 mol%. As described herein at page 10, line 18, to page 13, line 13, the content of carboxylic acid and lactone ring is calculated based on measuring the results of NMR peaks assigned to a lactone ring. The lactone-modified PVA of Watanabe et al contains 5 to 100 mol% of structural units derived from a lactone monomer, and thus does not overlap the presently-recited range of 0.02 to 0.4 mol%. Nor could one skilled in the art reading Watanabe et al in combination with Ikeda et al predict the above-discussed comparative results.

Tanaka et al, listed as "A" category, i.e., technical background, prior art, in the European Search Report of a corresponding European patent application, has been relied on by the Examiner as disclosing that a method for producing a ceramic molding comprising drying an aqueous kneaded material, to form granules, molding and sintering, are notoriously well-known in the ceramic arts. Interestingly enough, however, Tanaka et al is one of those prior art binders described in the specification in the paragraph bridging pages 3 and 4 as problematical. Comparative Example 6 of the above-discussed comparative data, employing PVA Polymer R, is an example of Tanaka et al. Comparative Example 6 is directly comparable to Example 6. The difference in results is striking, as shown in the above-discussed Tables 4A and 4B.

Tsuboi et al is also relied on for the same reasons as Tanaka et al is relied on. Tsuboi et al is directed to a sintering process of Mn-Zn ferrite, wherein the sintering pressure is higher than atmospheric pressure. There is no description therein, however, of a polymer binder.

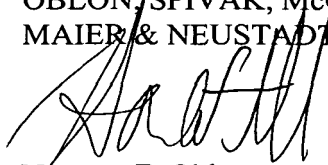
In addition to everything stated above, Ikeda et al disclose and suggest nothing with regard to using their PVA polymer as a ceramic-molding binder, or a compression-molding binder for ceramics.

For all the above reasons, it is respectfully requested that the rejections over prior art not be repeated.

All of the presently pending claims in this application are believed to be in immediate condition for allowance. Accordingly, the Examiner is respectfully requested to pass this application to issue.

Respectfully submitted,

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